

UNPUBLISHED

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Introduction

Kalamalka lake is by far the least productive of the mainstem lakes in the Okanagan drainage basin. It (also) is the only lake that experiences a very active calcium carbonate depositional cycle during the summer stratification season. The relationship between precipitation of calcium carbonate and low productivity has been covered quite extensively in the literature. Ohle (1952) suggested that suspended calcite, as an unuseable form of carbon, reduced photosynthesis in the Grosser Plön er See. Hutchinson (1957, p. 671), however, suggests that much of the colloidal and suspended carbonate observed by Ohle may have been in the form of organic complexes (carbmino carboxylic acid complexes) which are readily useable as a carbon source by algae (Smith et al, 1960). Suspended carbonate effectively adsorbs and complexes certain dissolved organic compounds, thus reducing the direct metabolic utilization of dissolved organics (Wetzel, 1968; 1970; 1972), and complexes and/or sorbs the metallic nutrients, especially iron (Schelske, 1962), as well as some of the macro nutrients such as phosphate (Wetzel, 1965; 1966; Cole et al, 1953), making them physiologically unavailable.

The present discussion does not, however, deal with any specific mechanism by which the lake has managed to remain so unproductive. A study much more detailed in scope than the monitor program completed during the 1971 season would be necessary to make this type of determination. Rather, this portion of the paper is intended to show that sufficient calcium carbonate is precipitated from the trophogenic zone each year to suggest that the calcite cycle has probably been an important factor in perpetuating the "unproductive" nature of this lake.

D.D. Williams (unpublished ^{manuscript} ~ 1972) The calcite cycle of Kalamalka L: Influence on lake productivity.

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Derivation of Carbonate Equilibria

The carbonate cycle was derived from measurements of temperature, pH, calcium and total alkalinity and the effects of changes in these on the equilibrium constants of carbonic acid. Strictly speaking, the values of these thermodynamic constants apply to carbonic acid in distilled water extrapolated to infinitely dilute concentrations, where, theoretically, there are no interactions between ions in solution (i.e. their activity = 1). Extremely dilute aqueous solutions, such as most rivers and lakes, can be considered for all practical purposes to closely approximate this condition. The major dissolved species can be looked upon as individual free ions and a chemical analysis of such waters reported in terms of the major ion content provides a satisfactory model of the internal economy of the solution. In such dilute solutions, where only the long-range electrostatic ionic interactions are of any significance, the activities of the various ionic species (accurate within a few per cent) can be obtained by multiplying the molal species concentration (determined by chemical analysis) by the ion activity coefficients derived from the Debye-Hückel theory. This approximation is fairly reasonable for waters of ionic strength less than 0.1 (Garrels & Christ, 1965). In more concentrated waters, the short-range electrostatic interactions between oppositely charged adjacent ions become increasingly significant (Garrels, Thompson & Siever, 1961) with the result that the effective, free ionic fraction of the ion in question is reduced by the formation of ion pairs.

The ionic strength of Kalamalka Lake water, calculated from the results of lake water analyses ~~done by the Western Water Quality Sub-division, DOE~~ (see Williams, M 1972) was 0.006. The effects of short-range interactions were, therefore, considered negligible. (Actual determinations by "successive approximations" of the contribution of the ion pairs CaSO_4^0 , CaHCO_3^+ , and MgCO_3^0 , showed that they removed less than 1% of the free ionic fraction of Ca^{2+} , CO_3^{2-} and HCO_3^- from solution). In consideration of this fact, the values of the activity coefficients for CO_3^{2-} (0.73) and HCO_3^- (0.94) used in the calculations were those determined by Walker, Bray & Johnson (1927) (see Garrels & Christ, 1965, Fig. 4.5). For similar reasons, the activity coefficient for Ca^{2+} (0.72) was obtained from Garrels & Christ (1965, Fig. 2.15).

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The carbonate equilibria were derived from the following relationships as outlined in Garrels & Christ (1965).

$$m_{\text{CO}_3^{=}} = \frac{m_{\text{HCO}_3^{-}} \cdot \gamma_{\text{HCO}_3^{-}} \cdot K_{\text{HCO}_3^{-}}}{a_{\text{H}^{+}} \cdot \gamma_{\text{CO}_3^{=}}} \quad (1)$$

$$m_{\text{Ca}^{2+}} = \frac{K_{\text{CaCO}_3}}{m_{\text{CO}_3^{=}} \cdot \gamma_{\text{Ca}^{2+}} \cdot \gamma_{\text{CO}_3^{=}}} \quad (2)$$

$$m_{\text{CO}_2(\text{aq})} = \frac{m_{\text{HCO}_3^{-}} \cdot \gamma_{\text{HCO}_3^{-}} \cdot a_{\text{H}^{+}}}{K_{\text{H}_2\text{CO}_3}} \quad (3)$$

$$\sum m_{\text{CO}_2} = \sum m_{\text{HCO}_3^{-}} + \sum m_{\text{CO}_3^{=}} + m_{\text{CO}_2(\text{aq})} \quad (4)$$

$$\text{IAP} = a_{\text{Ca}^{2+}} \cdot a_{\text{CO}_3^{=}} \quad (5)$$

The molal concentration of $\text{CO}_3^{=}$ ($m_{\text{CO}_3^{=}}$) was calculated from the analytical bicarbonate concentration, pH and the $K_{\text{HCO}_3^{-}}$ value at the in situ temperature extrapolated from the values determined by Harned and Scholes (1941). This was used in (2), to calculate from the solubility product relationship, the concentration of dissolved calcium for comparison with the analytical value of the calcium concentration as an indication of how much CaCO_3 might be in the colloidal or suspended form. It essentially indicates the degree of saturation of the water with respect to CaCO_3 , similar to (5). Values for K_{CaCO_3} at the in situ temperatures were extrapolated from those determined by Larson & Buswell (1942). This value for $m_{\text{CO}_3^{=}}$ was also used in the determination of the total CO_2 content of the water (4), and in the calculation of the IAP (ion activity product) in (5).

Using the values from Harned & Bonner (1945) for $K_{\text{H}_2\text{CO}_3}$ for the in situ temperature, the dissolved CO_2 ($m_{\text{CO}_2(\text{aq})}$) fraction ($\approx m_{\text{H}_2\text{CO}_3}$) of the total CO_2 content of the water was calculated.

Finally, using the calculated value for $m_{\text{CO}_3^{=}}$ (1) and the analytical

value for the calcium concentration ($\sum m_{Ca^{2+}}$), the ion activity product was determined for comparison with the equilibrium constant for calcite (K_{CaCO_3}) at the in situ temperature (Berner, 1965). This ratio, $IAP:K_{CaCO_3}$, gives an estimate of the degree of saturation of calcite in the water column, (i.e. $IAP:K_{CaCO_3} < 1$, the sample is undersaturated; $IAP:K_{CaCO_3} = 1$, the sample is saturated; $IAP:K_{CaCO_3} > 1$, the sample is supersaturated).

Several different approaches were attempted in calculating the quantity of calcium carbonate (calcite) precipitated from the lake each year. In each case, the calculations were limited to the 0 - 10 m volume, i.e. that amount of calcite precipitated through the 10 m contour.

A plot of the seasonal changes in the IAP: K_e ratio vs depth (Fig. 19), indicated that the calcite precipitated from the upper layers of the lake (epilimnion) was being re-dissolved in the hypolimnion. The sediment data corroborated this fact, with the calcium carbonate content of the sediments being approximately inversely proportional to water depth (St. John, ~~MS 1972~~^{unpublished}) (Fig. 17). The terrace deposits were composed almost entirely of calcite (85-90%), whereas the carbonate content of the deeper basin muds was only 10%. This would indicate the epilimnion to be the crystal generating source for the microcrystalline calcium carbonate. ~~Both these observations are supported by the work of Brunskill (1969) and Takahashi et al (1968)~~^{Similar} on meromictic, Fayetteville Green Lake, N.Y., which also actively precipitates calcium carbonate. Brunskill⁽¹⁹⁶⁹⁾ observed that crystals recovered from the surface waters tended to be sharply angled, whereas those from the deeper waters tended to be more stubby and rounded, suggesting that some re-solution ^{was possibly occurring.} Takahashi et al⁽¹⁹⁶⁸⁾ showed that the ^{13}C content of the surface sediments of the deep basin of the lake could be correlated with the dissolved, inorganic $^{13}\text{CO}_2$ of the surface water and was entirely different from that of the monimolimnion. Confining calculations to the volume above the 10 m contour has, hopefully, eliminated the complications introduced as a result of this re-solution process in the deeper waters:

From the work of Kemp and St. John (St. John, ~~MS 1972~~^{unpublished}) and Anderson (1972), excellent geochemical data exist for the terrace sediments. These data, in the absence of any direct measurements of the suspended crystal loads in the water column, permit a semi-empirical determination to be made of the quantity of calcite precipitated from the epilimnion each year, since the mean depth of the terraces (~10 m), approximates the mean depth of the thermocline over the summer stratification season.

The very unproductive nature of this lake has been well documented in works by Stockner^{et al.} (MS 1972) and ~~the present author~~ (Williams, MS 1972). In order to determine if the calcium carbonate cycle has been a major factor in "protecting" the lake from water quality deterioration since man settled in the Valley, as suggested by St. John (MS 1972), it is first necessary to determine the amount of calcium carbonate being precipitated each year. If indeed, complexing, sorption or co-precipitation phenomena are occurring as a result of calcite precipitation, thus

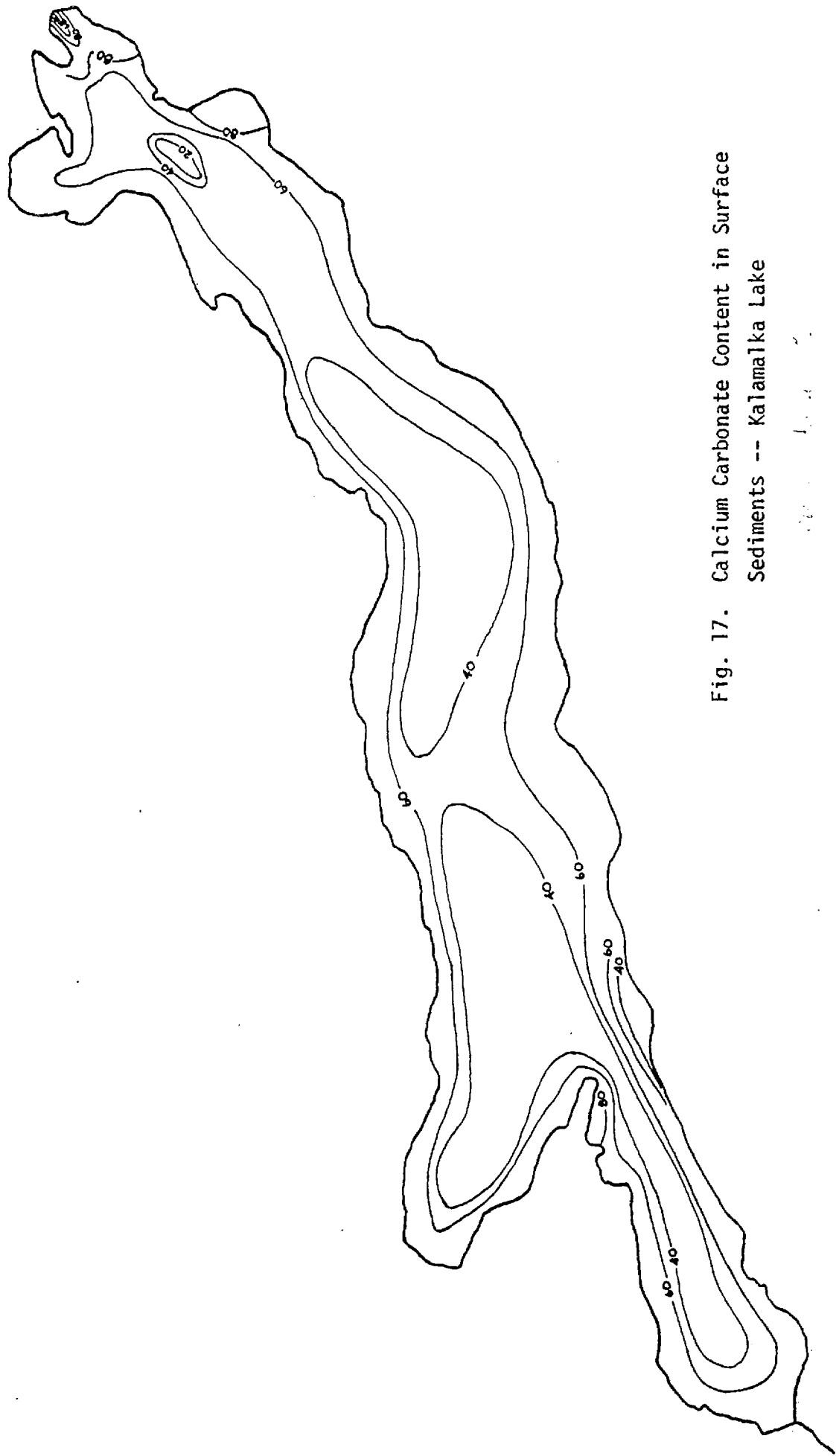


Fig. 17. Calcium Carbonate Content in Surface Sediments -- Kalamalka Lake

data from St. John (MS 1972)

decreasing the availability of organics, or such species as phosphate or iron to the algal community of the lake, the region of greatest importance insofar as the precipitation of calcite is concerned becomes the trophogenic zone, which can be approximated roughly by the surface 10 m.

The Precipitated Crystals

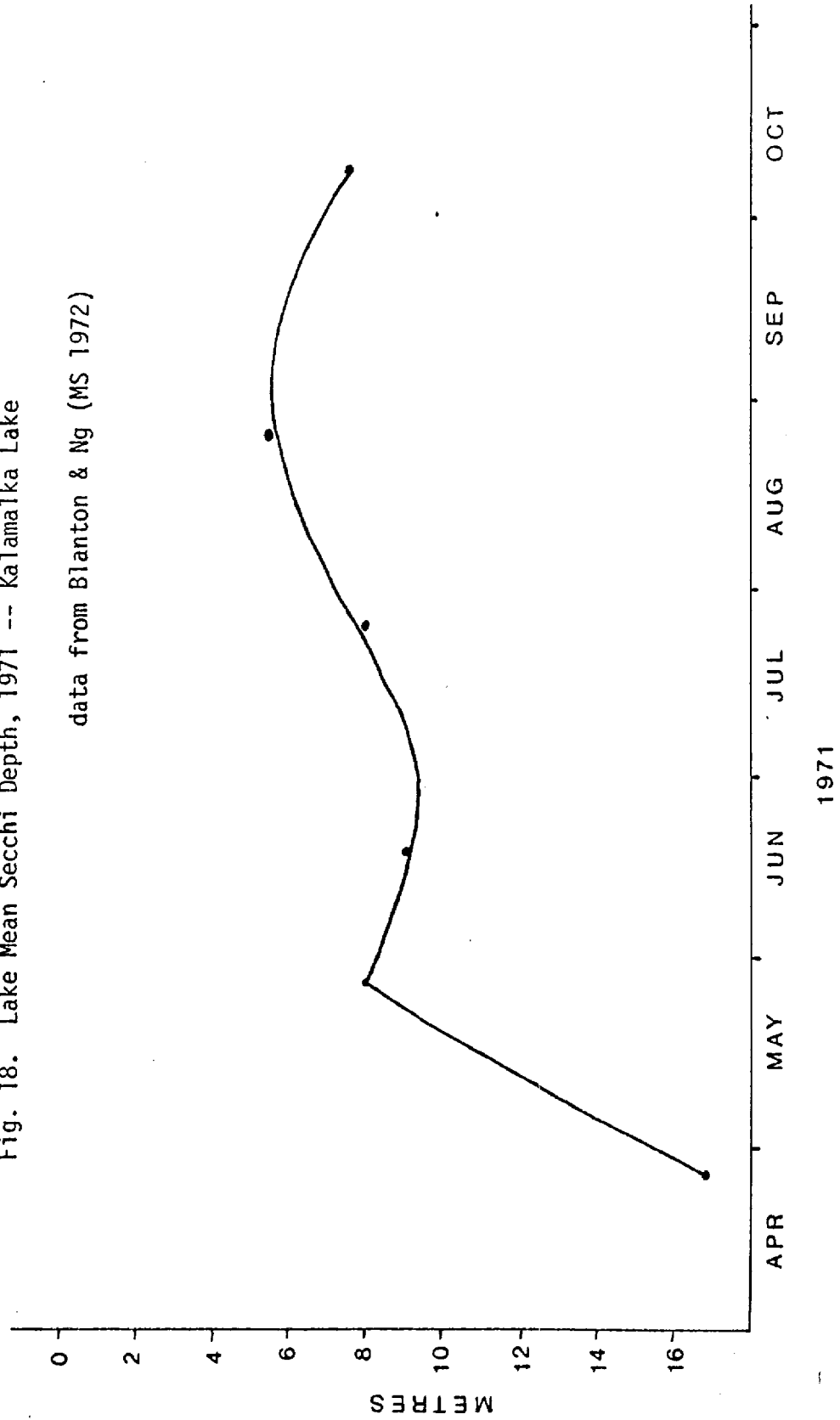
A drastic change in Secchi depth from 17 to 7 metres (Fig. 18) as well as a change in lake water appearance from dark blue to almost opalescent white in May, readily indicates the onset of crystal precipitation. Although no direct collection of suspended crystals from the water column was made, they were readily detected during the summer as precipitates on the casings of thermographs moored in the lake at a depth of 3 m (Blanton & Ng, ~~MS 1972~~), and on the glass slides of periphyton moorings at a depth of 2 m (Stockner, ~~MS 1972~~).

Crystals from Shipek and core samples taken of the terrace sediments were identified by micro-electron diffraction analysis as calcite, with hexagonal crystal structure and a size range of 0.5 to 5μ (^{Duncan personal communication} Radiochemistry Unit, CCIW). Their morphological features, examined under a scanning electron microscope (~~University of Toronto~~), indicated formation by nucleation processes at an original crystal surface and by dendritic growth with preferred orientations. Crystal aggregates were also observed.

These data agree reasonably well with the observations made by Brunskill (1969) on Fayetteville Green Lake. Measurements of crystals collected directly from the water column using sediment traps, and crystals from the sediment laminae, resulted in a mean nominal radius of 3μ , which closely approximates the crystal sizes observed in Kalamalka Lake. Crystal aggregates with diameters up to 60μ were also observed.

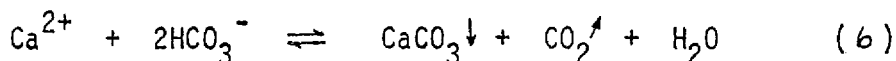
Fig. 18. Lake Mean Secchi Depth, 1971 -- Kalamalka Lake

data from Blanton & Ng (MS 1972)



Water Chemistry Data

The change in total CO_2 observed in the lake during the 1971 season is plotted in Fig. 20, indicating a decrease of approximately $0.18 \times 10^{-3} \sum m_{\text{CO}_2}$ from April through October (largely as a result of a decrease in $\sum m_{\text{HCO}_3^-}$). The effect was essentially limited to the surface 15 metres of the lake and agrees reasonably well with the seasonal distribution of temperature (Fig. 21). According to the equation,



a decrease of $0.18 \times 10^{-3} m_{\text{HCO}_3^-}$ requires the precipitation of $0.09 \times 10^{-3} m_{\text{CaCO}_3}$ and the evolution of $0.09 \times 10^{-3} m_{\text{CO}_2}$ either to the atmosphere or to be taken up by algae.

Calculated for the 0-10 metre volume of the lake (volume = $0.24 \times 10^9 \text{ m}^3$), this would result in the precipitation of $2.16 \times 10^9 \text{ g CaCO}_3$, assuming no solution of the crystals or loss to outflow.

The net change in the CO_2 system also requires a decrease of $0.11 \times 10^{-3} m_{\text{Ca}^{2+}}$ or the equivalent of 2.16×10^7 moles Ca^{2+} in the 0-10 m volume. A rough calcium budget worked out for the lake resulted in a loss of approximately 2.0×10^7 moles Ca^{2+} (Williams, unpublished data). The two estimates correspond extremely well.

Fig. 19 indicates a threefold increase in the apparent degree of supersaturation with respect to calcite of the surface few metres of the lake from June to September, with the maximum values occurring in August. In general, the isopleths follow the descent of the thermocline. Vertical cooling curves start appearing in late September (Fig. 21), and as the surface water cools, the saturation values start decreasing. In all probability, the lake fluctuates slightly around the saturation level during the winter months.

The increased saturation values can be almost entirely attributed to the changes in a_{CO_3} , which increased more than twofold from 1.15×10^{-5} in April to 2.60×10^{-5} in August, largely as the result of the effect of temperature on the equilibrium constant for HCO_3^- . m_{CO_2} also decreased in the surface waters over this period, from 6.00×10^{-5} to $2.80 \times 10^{-5} m_{\text{CO}_2}$. Removal of dissolved CO_2 increases the pH and thus favours the precipitation of CaCO_3 .

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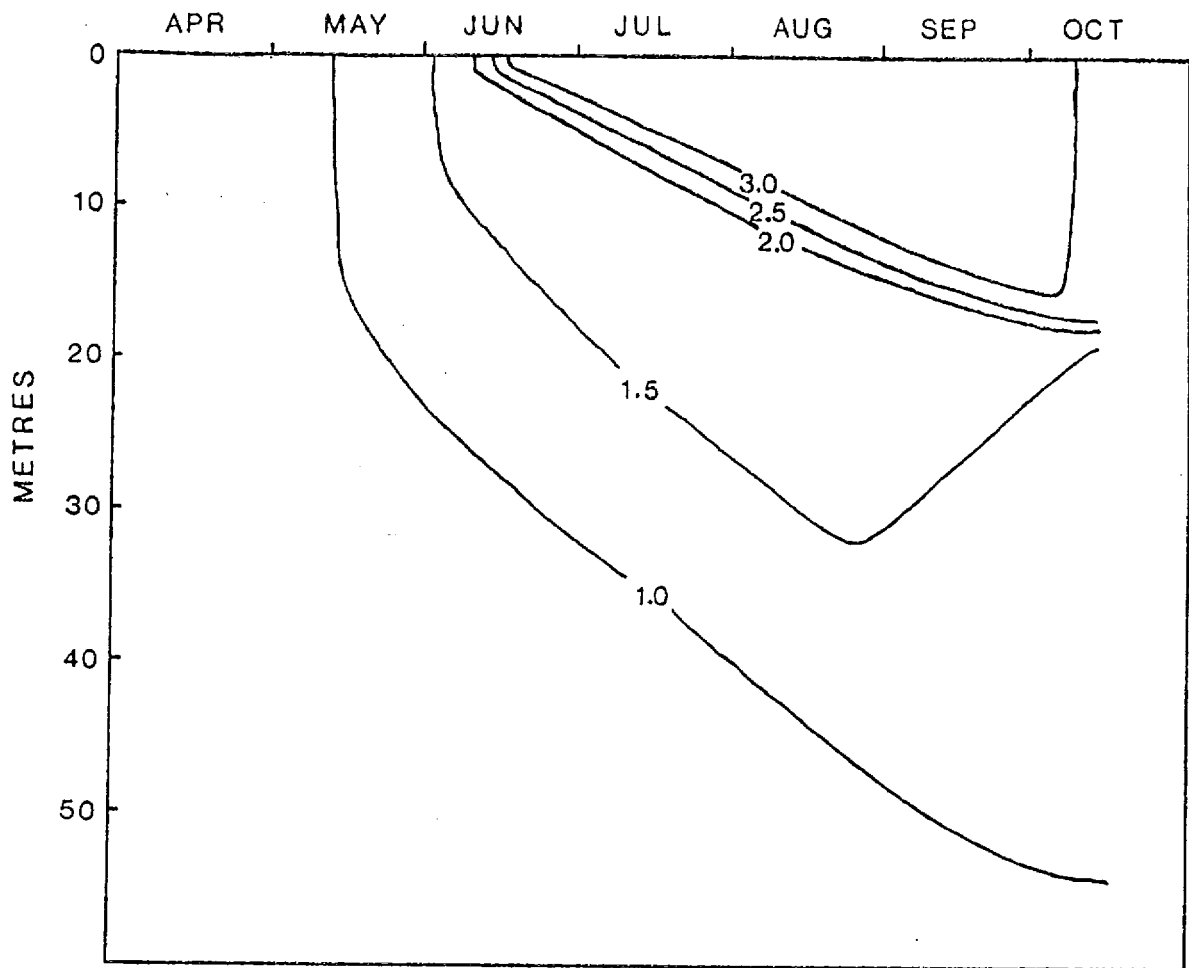


Fig. 19. Seasonal changes in the IAP:K_{CaCO3} Ratios, 1971 -- Kalamalka Lake

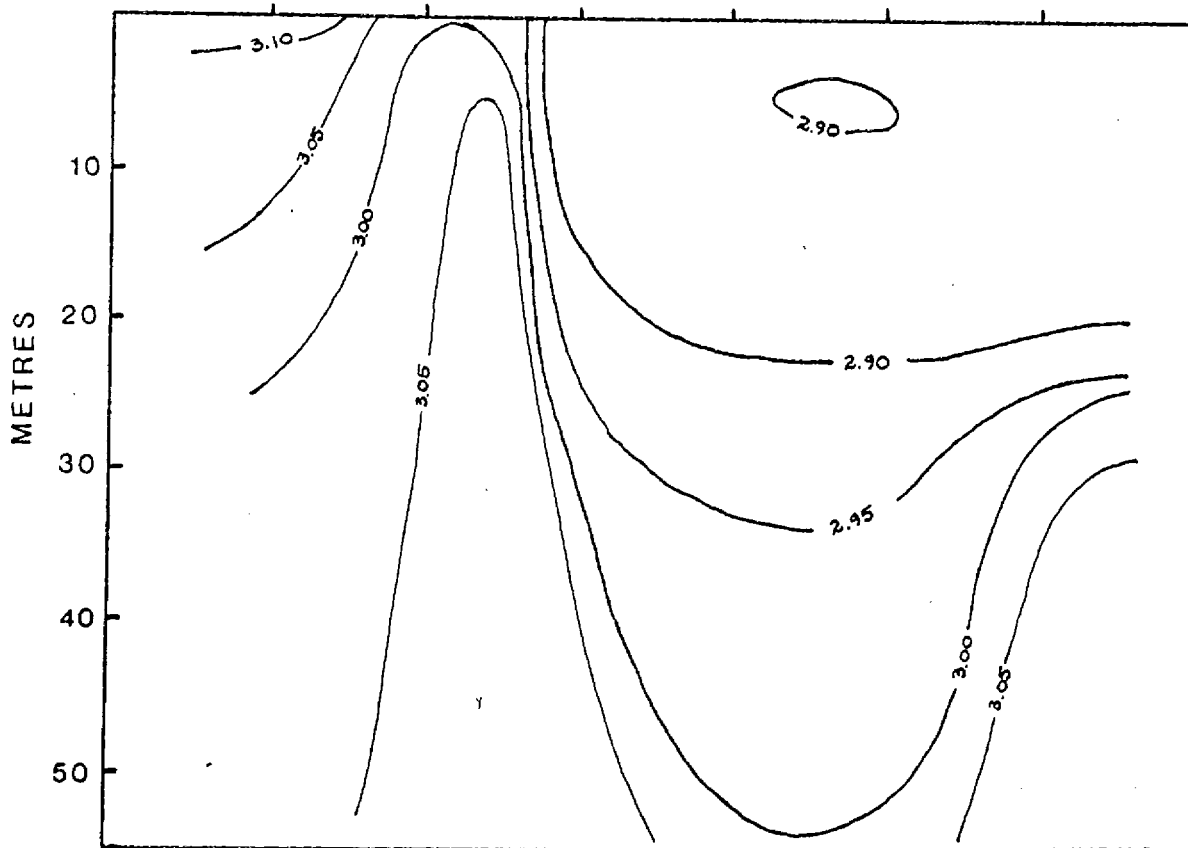


Fig. 20. Seasonal Distribution of ΣCO_2 (moles/litre x 10⁻³)

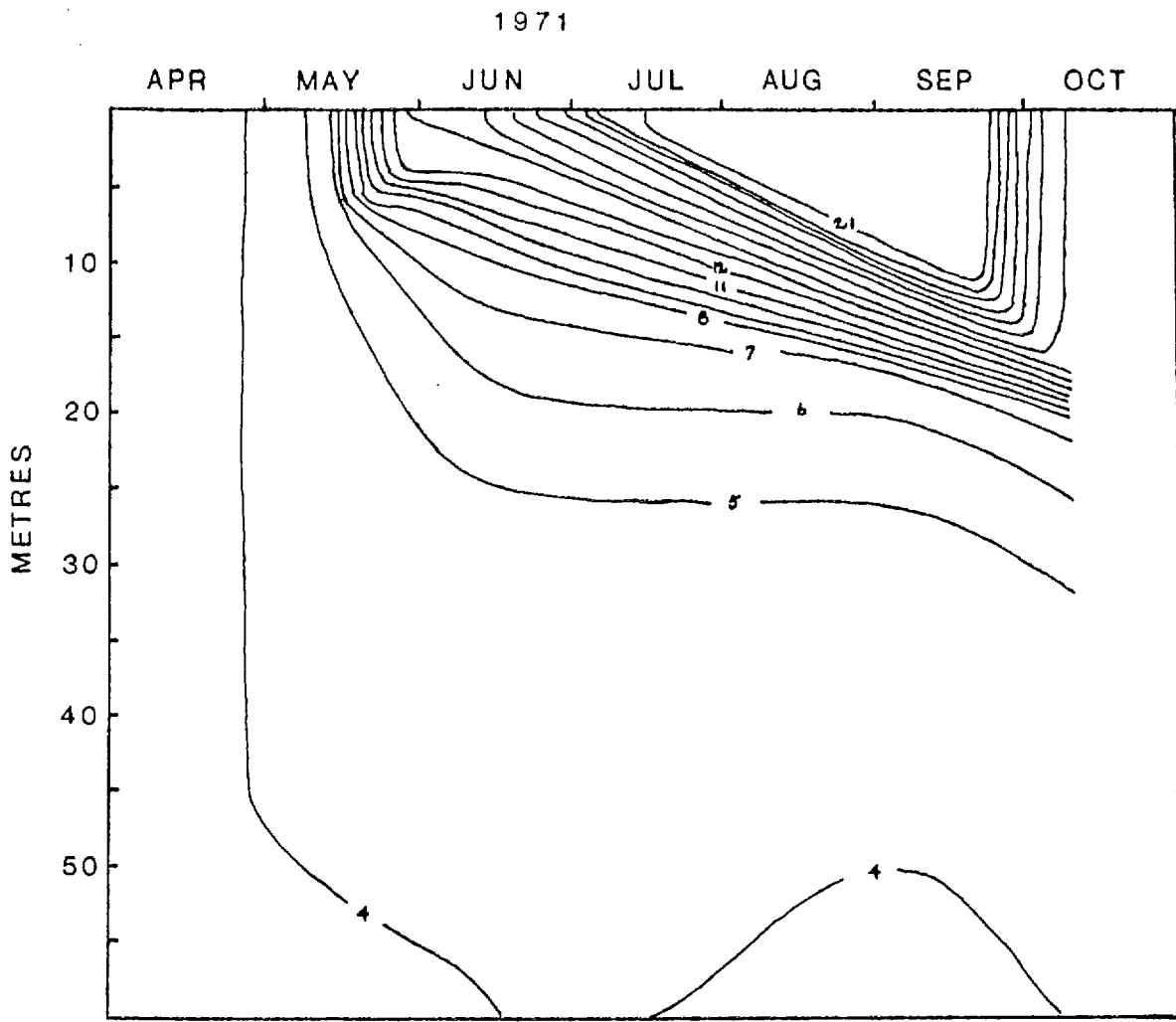


Fig. 21. Seasonal Temperature Isopleths -- Kalamalka Lake

Sediment Data

An attempt was made to correlate the results from the water chemistry analyses with the sediment data collected by Anderson, Kemp and St. John (St. John, ~~MS 1972~~; Anderson, ~~MS 1972~~).

Using sediment densities from Anderson (~~MS 1972~~) and mean water content for the uppermost sediment column (Kemp, unpublished data), the mean net annual accumulation of sediment (dry weight) for the lake was calculated by St. John as 413 g/m^2 lake surface/yr. This is equivalent to a sedimentation rate through the 10 m contour (area = $23 \times 10^6 \text{ m}^2$) of $9.5 \times 10^9 \text{ g/yr}$.

Kemp has determined a mean inorganic carbon content for the terrace sediments of 10.2% by weight (see St. John, ~~MS 1972~~). Using the total sedimentation rate calculated above, this is equivalent to $9.69 \times 10^8 \text{ g}$ (8.08×10^7 moles) inorganic carbon being deposited through the 10 m contour each year. If it is assumed that all this were in the form of calcium carbonate, then the total amount of calcite sedimented from the 0 - 10 m volume amounts to $8.08 \times 10^9 \text{ g/yr}$. About 10% of this is of terrigenous origin (St. John, MS 1972) which results in $7.3 \times 10^9 \text{ g}$ being precipitated from the water in the lake.

St. John (op. cit) has determined a mean Ca^{2+} content for the terrace sediments (as CaO) of 49.8% by weight. Using similar considerations as above for inorganic carbon, the calculations result in $8.45 \times 10^9 \text{ g CaCO}_3$ being precipitated through the 10 m contour. Allowing for the 10% terrigenous fraction, results in $7.6 \times 10^9 \text{ g/yr}$ originating from the water column. The two results show strikingly good agreement, suggesting that the assumption made above was probably quite valid. The slight discrepancy that does exist could readily be attributed to sample variability and/or differences produced by the analytical methods used for measuring the two parameters.

Considering that the water chemistry data are based on only one year's observation (April to October), whereas the sediment data are based on the mean of a hundred years' accumulation of sediment, the correlation between the two sets of results is also quite reasonable. The apparently higher sedimentation rate for calcite obtained from the sediment data ($7.5 \times 10^9 \text{ g/yr}$) as compared to that obtained from the water chemistry ($2.2 \times 10^9 \text{ g/yr}$) could also be attributed to the fact that a total sedimentation rate based on the results for the deeper basin muds was used for the calculations. Considering the morphometry of this lake, it is questionable (in the author's opinion) whether this value can be validly extrapolated over the entire surface.

Fig. 19 indicates that the water is still supersaturated with respect to

calcite in October. The fraction of this suspended calcite that would probably be precipitated from the water column before the surface waters return to saturation, was not included in the calculations. This would obviously tend to increase the sedimentation rate obtained from the water chemistry data.

It is probable that most of the calcium carbonate precipitated from this lake originates by inorganic, as opposed to biogenic processes, with the main initiating (and terminating) factor being the effect of temperature on the equilibria constants for carbonic acid and the activity product constant for CaCO_3 . Photosynthesis probably functions only in a minor secondary role.

The onset of crystal precipitation in late May, as evidenced by the change in both Secchi depth and water appearance, coincides with the beginning of stratification and formation of the thermocline. Epilimnion heating rates in the order of $250\text{-}350 \text{ g.cal/cm}^2/\text{da}$ persist throughout the period April to mid-June (Blanton & Ng, MS 1972). The IAP: K_{CaCO_3} ratio increases throughout the summer, with the isopleths generally following the descent of the thermocline. Total dissolved CO_2 starts decreasing in mid-May. The high values for IAP: K_{CaCO_3} which persist throughout the stratification period may indicate that the surface waters were being constantly replenished with CO_2 (perhaps by mixing and warming of water in the thermocline, see Brunskill, 1969). It may also indicate that the turbulence in the epilimnion was tending to keep the crystals in suspension. This would also explain, in part, the relatively lower value for calcite precipitation calculated from water chemistry data (April - October) as opposed to the value obtained from sediment data.

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Although no direct studies were made to determine if the very unproductive nature of Kalamalka Lake could be linked with the active calcium carbonate cycle, the calculations show that the quantity of calcite precipitated from the trophogenic region each year is sufficient enough to suggest, at least, that it could be acting as a "protective mechanism" in this lake.

Crystal Settling Velocities

Although it was considered unreasonable to assume that crystal sedimentation rates based on Stokes' Law could be calculated for the region of the lake above the thermocline, it was thought that such would not be the case for the hypolimnion. This region of the lake does not experience the turbulence of the epilimnion, which would, in general, tend to keep the crystals in suspension. In addition, any error associated with the effect of changing temperature on water viscosity (which can be quite considerable, ~~but~~) is drastically reduced by the isothermal nature of the hypolimnion.

The effect of temperature on the carbonate equilibria constants appears to be the initiating factor for calcite precipitation in this lake (see discussion below). Since the temperature fluctuations below 25 m were virtually negligible (Fig. 21), it was assumed that the apparent supersaturation of calcite in the hypolimnion below this depth was the result of precipitation from the layers of the lake above. Consequently, the rate of descent of the $IAP:K_e = 1.5$ isopleth was used as being representative of the "settling velocity" of the calcite crystals. This resulted in a value of 2.89×10^{-4} cm/sec. Substituting this value into the Stokes' equation,

$$v = \frac{2}{9} \frac{(\rho_1 - \rho_2)gr^2}{\eta} \quad (7)$$

where, ρ_1 = density of calcite (2.71 g/cm³)

ρ_2 = density of lake water (1.00 g/cm³)

g = acceleration due to gravity (980 cm/sec²)

v = particle settling velocity (2.89×10^{-4} cm/sec)

η = viscosity of water (0.016 at 4.5°C) (g/cm.sec) = 1 poise

r = particle radius

resulted in a particle size of approximately ^{1.1} 2μ . This is ~~in the middle~~ ^{at least within} of the crystal size range determined by electron diffraction.

It would be interesting to compare the approximated settling velocity with that observed using actual crystals collected from the water column and settling tube techniques.

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NOTE: References marked with an asterisk (*), were reports submitted to the Canada-British Columbia Okanagan Basin Study Committee in fulfilment of tasks funded under the Canada-British Columbia Okanagan Basin Agreement.